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# Antarctic-wide array of high-resolution ice core records reveals pervasive lead pollution began in 1889 and persists today

Received  
14 May 2014Accepted  
7 July 2014Published  
28 July 2014J. R. McConnell<sup>1</sup>, O. J. Maselli<sup>1</sup>, M. Sigl<sup>1</sup>, P. Vallelonga<sup>2</sup>, T. Neumann<sup>3</sup>, H. Anshütz<sup>4</sup>, R. C. Bales<sup>5</sup>, M. A. J. Curran<sup>6</sup>, S. B. Das<sup>7</sup>, R. Edwards<sup>8</sup>, S. Kipfstuhl<sup>9</sup>, L. Layman<sup>1</sup> & E. R. Thomas<sup>10</sup><sup>1</sup>Desert Research Institute, USA, <sup>2</sup>Centre for Ice and Climate, University of Copenhagen, Denmark, <sup>3</sup>NASA Goddard Space Flight Center, USA, <sup>4</sup>Norwegian Geotechnical Institute, Norway, <sup>5</sup>University of California, Merced, USA, <sup>6</sup>Australian Antarctic Division, Australia, <sup>7</sup>Woods Hole Oceanographic Institution, USA, <sup>8</sup>Imaging and Applied Physics, Curtin University, Australia, <sup>9</sup>Alfred-Wegener-Institut, Helmholtz-Zentrum für Polar- und Meeresforschung, Germany, <sup>10</sup>British Antarctic Survey, UK.

Correspondence and  
requests for materials  
should be addressed to  
J.R.M. (Joe.  
McConnell@dri.edu)

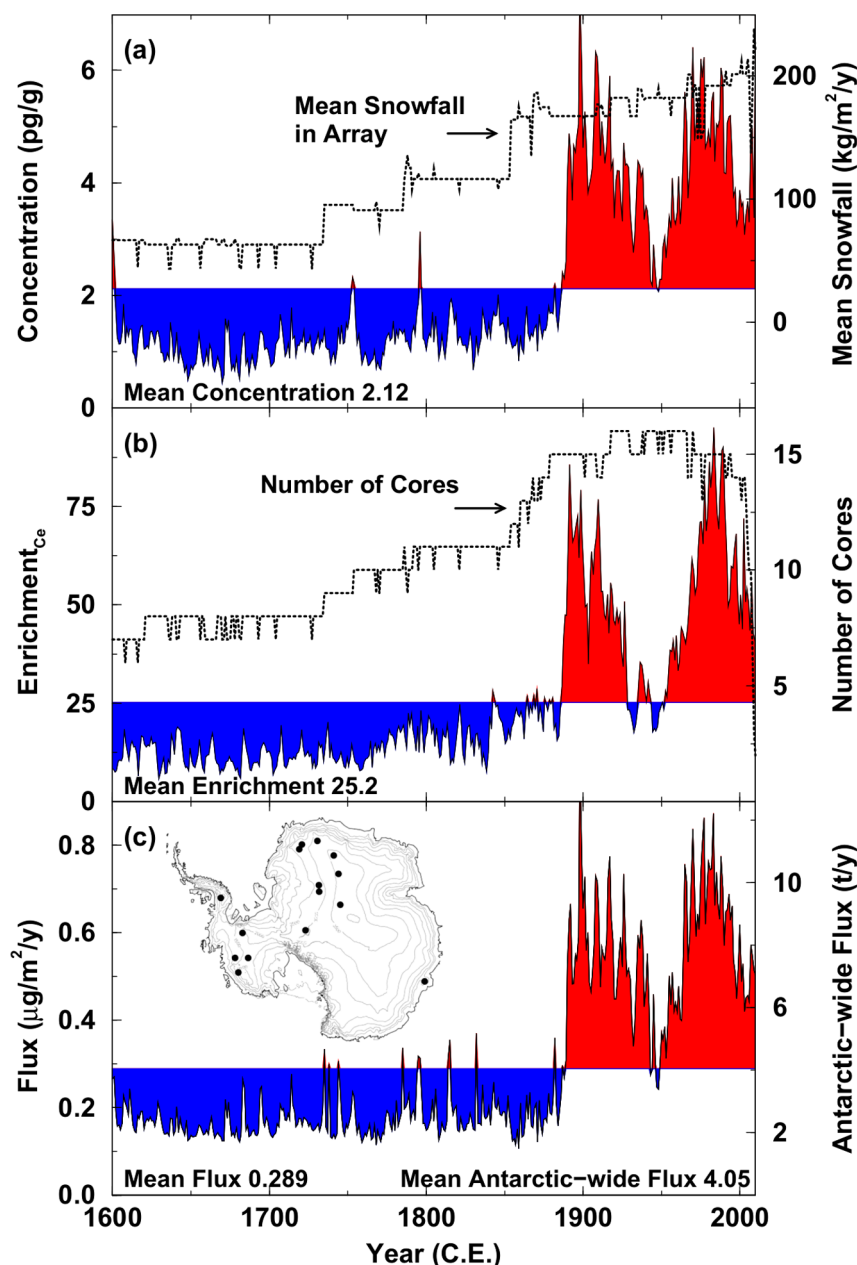
Interior Antarctica is among the most remote places on Earth and was thought to be beyond the reach of human impacts when Amundsen and Scott raced to the South Pole in 1911. Here we show detailed measurements from an extensive array of 16 ice cores quantifying substantial toxic heavy metal lead pollution at South Pole and throughout Antarctica by 1889 – beating polar explorers by more than 22 years. Unlike the Arctic where lead pollution peaked in the 1970s, lead pollution in Antarctica was as high in the early 20<sup>th</sup> century as at any time since industrialization. The similar timing and magnitude of changes in lead deposition across Antarctica, as well as the characteristic isotopic signature of Broken Hill lead found throughout the continent, suggest that this single emission source in southern Australia was responsible for the introduction of lead pollution into Antarctica at the end of the 19<sup>th</sup> century and remains a significant source today. An estimated 660 t of industrial lead have been deposited over Antarctica during the past 130 years as a result of mid-latitude industrial emissions, with regional-to-global scale circulation likely modulating aerosol concentrations. Despite abatement efforts, significant lead pollution in Antarctica persists into the 21<sup>st</sup> century.

Lead is a toxic heavy metal with the potential to damage ecosystems. Low background atmospheric concentrations together with well-known and often distinct isotopic characteristics of industrial sources make lead an ideal tracer of industrial pollution<sup>1–7</sup>. Historical records of preindustrial to present lead concentrations and lead isotope ratios measured in ice cores have been reported for Antarctica, but these studies included cores and snow pits from only three coastal sites<sup>6,8–11</sup> (Supplementary Fig. S1), each with relatively few (generally <100) discrete measurements based on irregular and discontinuous sampling spanning different time periods (Supplementary Fig. S2). These records enabled limited evaluation of the onset, severity, and changes in industrial lead concentrations in Antarctica but revealed little about deposition fluxes, changes in atmospheric transport and in particular, about the extent of contamination in the vast interior region, or the efficacy of recent emission abatement efforts in reducing lead pollution in Antarctica.

Here we used continuous measurements (Methods Summary) of lead concentration, enrichment, and deposition flux in an array of 16 recently collected ice cores from widely spaced coastal and interior sites (Fig. 1) to develop a year-by-year understanding of the history of lead pollution throughout Antarctica since 1600 C.E. and extending into the 21<sup>st</sup> century. Results show that industrial lead pollution was pervasive in Antarctica as early as the late 1880s likely originating from a single Australian source and that lead contamination persists in the early 21<sup>st</sup> century. Changes during the past 410 years were surprisingly similar across all of Antarctica (Supplementary Fig. S4), indicating that anthropogenic lead is well mixed in the Antarctic atmosphere and deposition of this toxic heavy metal is ubiquitous across the continent.

## Results and Discussion

Atmospheric aerosol concentrations are reflected in precipitation so measurements in well-dated ice cores can provide a proxy of historical changes in the atmosphere<sup>1,2,4–7</sup>. Enrichment also is a widely used indicator of



**Figure 1 | Composite ice core records of lead in Antarctica from 1600 to 2010 C.E.** Records are in annual resolution. Areas shaded in blue and red indicate time periods where the lead value is below or above the 1600 to 2010 mean, respectively, highlighting the marked changes before and after industrialization in the mid- and low-latitudes. (a) Lead concentration and the mean snowfall each year for the composite record. (b) Enrichment of lead relative to the dust proxy cerium<sup>28</sup> and number of cores included in the composites each year. (c) Lead flux measured in the array after adjustment to account for site-specific differences in annual snowfall (left axis) and estimated Antarctic-wide flux (right axis). Integration of the red area indicates that ~610 t of industrial lead have been deposited in Antarctica during the past 130 years. Inset map shows the locations of the ice cores used in this study. Inset map was generated using Panoply Data Viewer software; Schmunk, Robert B. (2014). Panoply Data Viewer (version 4.0.1) [Software]. Available from <http://www.giss.nasa.gov/tools/panoply/>.

pollution and is the ratio of measured lead-to-dust concentration in the ice core divided by the concentration ratio found in mean crustal material or sediment<sup>12</sup>, with values above 10 clearly indicating anthropogenic lead input<sup>13</sup>. We developed composite records for concentration and enrichment from the Antarctic array by taking the geometric mean of annual average values measured in each ice core.

Lead concentrations measured in the array generally agree with previous coastal site studies<sup>6,8–11</sup> (Supplementary Fig. S2) but show that lead pollution extended into the vast interior region even during the late 19<sup>th</sup> century. Concentrations (five-year averages;

Supplementary Fig. S5) gradually increased from ~0.6 pg g<sup>-1</sup> in 1650 C.E. to ~1.8 pg g<sup>-1</sup> in 1885, likely reflecting a relatively minor increase in lead emissions from early mining and smelting activities in the mid- and low-southern latitudes. Antarctic concentrations increased very rapidly after 1885 reaching a maximum of 5.4 pg g<sup>-1</sup> by 1900. Concentrations remained high until the late 1920s, with a temporary low during the Great Depression (~1932) and again at the end of WWII (~1948) when concentrations dropped back to mid-19<sup>th</sup> century levels. Concentrations increased rapidly to 5.7 pg g<sup>-1</sup> by 1975 and remained elevated until the mid-1990s. Concentrations during the early 21<sup>st</sup> century were ~3.7 pg g<sup>-1</sup> lower



than the peak 20<sup>th</sup> century concentrations but well above background levels before the start of the Industrial Revolution.

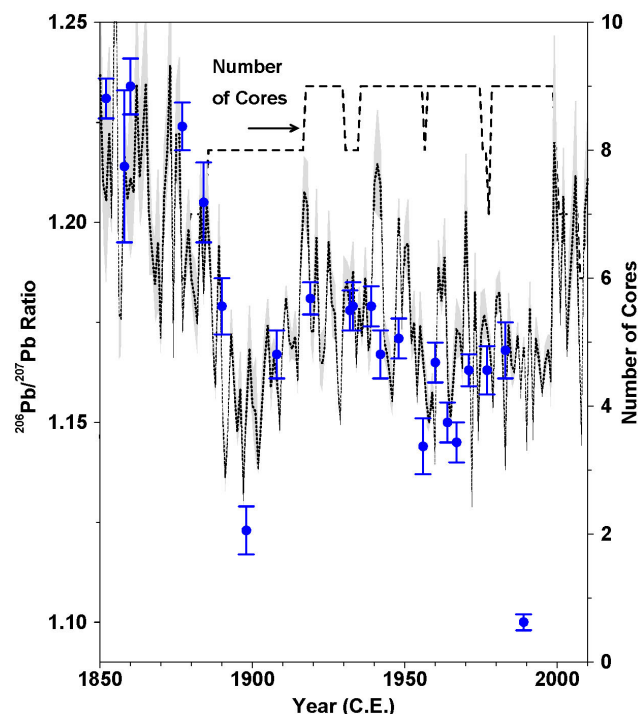
Lead enrichment (Fig. 1; Supplementary Fig. S5) generally followed changes in concentration. Five-year averaged enrichment values ranged from a background of  $\sim 10$  consistent with previous measurements<sup>13</sup> to  $\sim 70$  in 1895 and  $>85$  in 1982. Enrichment declined in the early 21<sup>st</sup> century but as with concentration, fluctuated between  $\sim$ four-to-five times background levels.

Deposition flux also is a widely used proxy for atmospheric aerosol concentration, but quantitative lead flux records previously have not been reported for Antarctica because contiguous lead measurements are required, as well as annual snowfall amounts. Flux differs from concentration because wet and dry deposition processes vary between sites with different snowfall rates. Using wet and dry deposition parameters determined from the array (see Methods section), we removed the snowfall-dependent component in each record before combining the records to create array and continent-wide composites of lead flux. Background flux for the continent-wide composite averaged  $2.4 \text{ t y}^{-1}$ , but by 1890 fluxes had increased more than four-fold, reaching a maximum five-year average of  $10.6 \text{ t y}^{-1}$ . Fluxes followed the same general trend as lead concentrations, with a minimum of  $4.0 \text{ t y}^{-1}$  in 1947, followed by a rapid increase back to 1890 levels before decreasing to  $7.0 \text{ t y}^{-1}$  by the early 21<sup>st</sup> century – still nearly three times background levels. Integrating the annual Antarctic-wide fluxes above the background shows that  $\sim 660 \text{ t}$  of industrial lead have been deposited on continental Antarctica, with  $\sim 140 \text{ t}$  already deposited by 1911 when Amundsen arrived.

Quiescent volcanic emissions contribute to background levels of lead<sup>14</sup>, leading to preindustrial enrichment factors of  $\sim 10$  (Fig. 1b). Consistent with previous studies<sup>15</sup>, short-term (one to two year) increases in lead concentrations such as in 1753 and 1796 (Fig. 1a) are linked to increased continental dust concentrations, not volcanic eruptions, and so do not coincide with short-term enrichment anomalies. Only six of the 17 years with the highest lead fluxes prior to 1889 (Fig. 1c) coincide with known volcanic eruptions<sup>16,17</sup>.

Isotopic ratios contain information on lead provenance, provided that ratios of possible sources are known and sufficiently distinct<sup>4,5</sup>. While background isotopic ratios may vary between sites and through time because of variations in continental dust and volcanic lead inputs, ratios will be similar for periods when all cores are significantly impacted by the same industrial pollution source. Continuous, semi-quantitative measurements of  $^{206}\text{Pb}/^{207}\text{Pb}$  isotopic ratios made on nine of the 16 ice cores used in this study (Supplementary Fig. S1) were combined to create a composite record from 1850 to 2010 C.E. (Fig. 2).

The isotopic composite record reveals distinct changes in the sources of lead reaching Antarctica (Fig. 2), and changes were observed across the continent suggesting that a single emission source was responsible for polluting Antarctica and possibly large regions of the SH at the end of the 19<sup>th</sup> century and remained a significant source throughout the 20<sup>th</sup> century. The composite record of lead  $^{206}\text{Pb}/^{207}\text{Pb}$  isotopic ratios (Fig. 2) shows a gradual decline from 1850 to 1886, with values ranging from 1.18 to 1.22 (five-year averages; Supplementary Fig. S6) – coincident with similar gradual increases in lead concentration and enrichment (Fig. 1). A pronounced decline began in 1887, culminating in a distinct minimum of  $<1.13$  in 1891 and generally low ratios from 1890 to 1905. The Australian Broken Hill lead/silver ore body is characterized by particularly low lead  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios of  $1.0407 \pm 0.0006^3$ , and the Port Pirie smelter in Australia was established in 1891 to process Broken Hill ores. The low isotopic ratios observed in Antarctic ice suggest that Broken Hill was the source and Port Pirie was the point of emission for rapidly increasing lead fluxes and enrichment after 1891<sup>8</sup>.  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios gradually increased through 1912 and then stabilized within a range of 1.17 to 1.20 until 1956. A broad decline in the isotopic ratios followed, reaching a minimum of 1.15 in 1995 in



**Figure 2 |** Composite record of semi-quantitative  $^{206}\text{Pb}/^{207}\text{Pb}$  isotopic ratios from this study (geometric average) compared with previously measured values from Law Dome<sup>6</sup> (blue, error bar indicates standard error). We consider the measurements semi-quantitative because no calibration specific for lead isotopic ratios was made during our continuous measurements. Grey shading indicates the standard error in the composite mean values. The number of ice cores included in the composite incorporates measurements from two coastal and seven inland sites spanning both East and West Antarctica. Temporal evolution of the Antarctic-wide isotopic composite record also is consistent with discrete measurements from the previously studied coastal sites (see also Supplementary Fig. 6 online), with only small differences in magnitude during the late 19<sup>th</sup> and early 20<sup>th</sup> centuries.

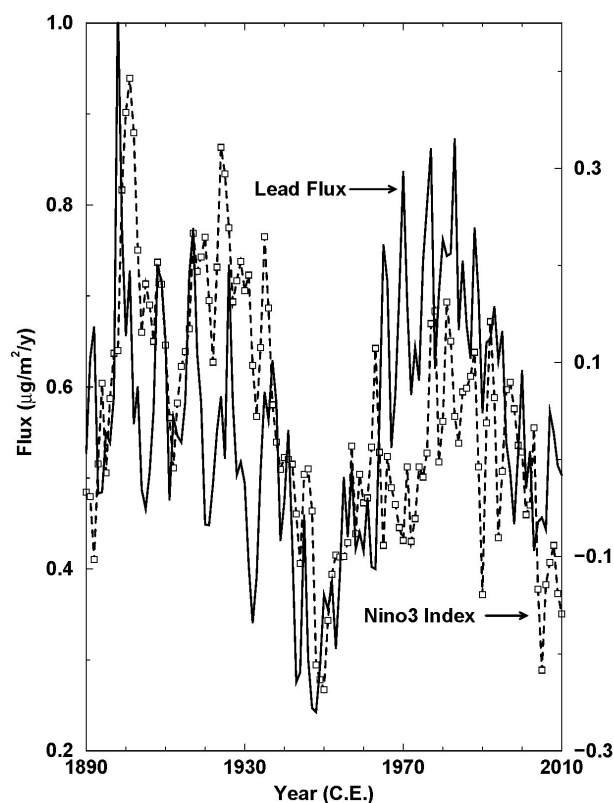
parallel with high concentration and fluxes and likely attributed to alkyl-lead from leaded-gasoline emissions<sup>8</sup>. Although lead ores in southern Africa feature similar lead isotopic compositions to those of Broken Hill, statistical data demonstrate that Pb emissions from southern Africa were consistently insignificant with respect to those from Australia. Lead production statistics from southern Africa prior to 1938 are limited<sup>18</sup>, however after this time Pb ore production was recorded as a mere 25 kt compared with 274 kt produced in Australia<sup>19</sup>. Lead was totally eliminated from gasoline in the largest and southernmost countries of the SH after this time: New Zealand (1996), Brazil (1997), Argentina (1998), Chile (2001), Australia (2002), and South Africa (2006)<sup>20,21</sup>. This is seen in the isotopic ratios which increased rapidly after 1996 and remained high, in the range 1.18 to 1.20 since, corresponding to a period of generally lower concentration and enrichment (Fig. 1).

Ice core aerosol records reflect changes in emissions as well as atmospheric circulation and transport processes; with no sources in Antarctica, industrial lead provides an ideal indicator for general circulation modeling studies by providing a highly resolved record of past changes in long-range aerosol transport from mid- to low-latitudes to the high southern latitudes. Indeed, lead deposition in Antarctica is highest in spring each year when long-range transport is most vigorous (Supplementary Fig. S3). The El Niño-Southern Oscillation (ENSO) is the variation in equatorial Pacific sea surface temperature that induces regional-scale changes in atmospheric circulation in the high southern latitudes. Several researchers have



reported links between ENSO and aerosol deposition flux in Antarctica, although typically the comparison is limited to decadal timescales due to limitations in temporal resolution of the ice cores<sup>22,23</sup>. Spectral analysis<sup>24</sup> of our composite of eight records from high snowfall sites reveals that inter-annual ENSO variations also influence lead deposition in Antarctica (Methods Summary; Supplementary Fig. S8). There is a consistent and significant variability in the lead flux with periods of ~3.5, 4.0, and 5.5 years before and after industrialization (Supplementary Fig. S8) – with the former linked to long-range transport of volcanic lead and dust from the mid-latitudes and the latter mainly to transport of industrial lead.

Comparisons to ENSO (NINO3 Index<sup>25,26</sup>; Fig. 3) indicate that much of the observed multi-decadal variability in Antarctic lead records during the past 120 years also may be attributed to changes in atmospheric transport, suggesting that regional-to-global scale circulation plays an important role in modulating aerosol concentrations and climate at multi-decadal timescales. Despite availability of statistics for mining and leaded fuel consumption in the SH, without detailed knowledge of the lead emissions that these activities produced, it is difficult to separate quantitatively the influence of changes in emissions and atmospheric transport to the resultant lead-deposition flux. However, our array of continuous, high-resolution lead records demonstrates unambiguously the marked impacts of SH industrial emissions on the Antarctic environment decades before the arrival of polar explorers in December 1911 and that industrial pollution of Antarctica persists today.



**Figure 3** | Comparison between 21-year running means of the composite lead flux from the eight ice cores individually dated using annual layer counting; Supplementary Fig. 8a) and a reconstruction of the ENSO Index (NINO3). Similarities between multi-decadal ENSO variability and lead flux during the industrial era (post-1890) indicate that changes in long-range transport in addition to emissions may contribute to the observed variations in lead deposition over Antarctica.

## Methods

This study is based on continuous measurements of lead, cerium, and other elements and chemical species in an array of 16 recently collected ice cores from widely spaced coastal and interior sites (Supplementary Fig. S1; Supplementary Table S1). Measurements were made using a well-established continuous ice core analytical system<sup>27</sup> with an effective depth resolution of ~0.01 m yielding between ~4 and >100 samples per year (Supplementary Fig. S3) and the ice core records dated using annual-layer counting and volcanic synchronization (Supplementary Table S1)<sup>16,17</sup>. We calculated enrichment from the ratio of annual lead-to-cerium<sup>28</sup> concentrations divided by the mass ratio of lead-to-cerium in mean sediment, 0.229<sup>12</sup>. Both concentration and enrichment composite records are displayed as the geometric mean<sup>29</sup> without any correction for differences in site average snowfall.

Lead measurements were performed on an ICP-MS (Thermo Scientific Element 2); a double focusing sector field mass spectrometer with argon plasma source. The ice core melt system that feeds into the ICP-MS is acid cleaned twice daily, and the ICP-MS is housed in a class-100 clean room to minimize potential lead contamination. Procedural blanks showed extremely low background levels of <sup>208</sup>Pb, typically 0.052 (±0.005, 2σ) pg/g, much lower than the lowest concentrations recorded in the Antarctic ice cores. Custom made lead standards (Inorganic Ventures, DRI-CAL-9C) were used for <sup>208</sup>Pb calibrations. Accuracy of individual <sup>208</sup>Pb measurements was calculated as being within 1 pg/g with typical precision of less than 0.5%.

The isotopic ratio measurements are described as semi-quantitative since the minor lead isotopes (<sup>206</sup>Pb and <sup>207</sup>Pb) were not calibrated with independent standards but assumed to have the same detection sensitivity as the most abundant isotope (<sup>208</sup>Pb) and then calibrated according to their detection response relative to <sup>208</sup>Pb. Relative detection sensitivity of the ICP-MS technique depends upon the differences in ionization potential of the different isotopes as well as any mass-biases within the magnetic field collection system – both of which may contribute to a disparity between the true and measured isotopic ratio<sup>30</sup>. In this study, the <sup>206</sup>Pb/<sup>207</sup>Pb ratio measured in the calibration standards was 1.086 (±0.003, 2σ) which is within 0.6% of the isotopic natural abundance ratio (1.0934)<sup>31</sup> demonstrating the small differences in the relative detection efficiencies of the two isotopes in this system. Comparison between the isotopic ratio records from all ice cores in the Antarctic array reveals that the variability in the absolute ratio between sites (between 1 and 10% of the mean value) overwhelms any measurement variability caused by the semi-quantitative calibration technique used here. Indeed, comparison between the Antarctic array isotopic ratio record and that from individual sites from other studies (Extended Data Fig. 4) confirms that any sensitivity corrections omitted by the semi-analytical approach are negligible compared with inter-site isotopic ratio variability.

To minimize snowfall-dependent differences in fluxes within the array, we mapped individual flux records to the average Antarctic snowfall rate (166 kg m<sup>-2</sup> y<sup>-1</sup>)<sup>32</sup> using wet and dry deposition parameters determined from the array before averaging them to create both array and continent-wide composites (Fig. 1). Wet and dry deposition parameters were determined for background (1600 to 1850 C.E.) and polluted (1890 to 2000 C.E.) flux conditions using the 11 ice core records that extended through both time periods (Supplementary Fig. S7), exploiting the large range of snowfall rates included in the array (e.g., >690 kg m<sup>-2</sup> y<sup>-1</sup> at Gomez to <27 kg m<sup>-2</sup> y<sup>-1</sup> at NUS7-5 [Supplementary Table S1]). The snowfall correction used for each ice core was:

$$\text{Flux}_{\text{corrected}} = \text{Flux}_{\text{original}} - m_{p/i} (\text{AveAccum}_{p/i} - 166) \quad (1)$$

where  $m_{p/i}$  is the slope of the preindustrial/industrial accumulation, flux relationship (Supplementary Fig. S7) and  $\text{AveAccum}_{p/i}$  is the average snowfall at that site during the preindustrial/industrial period. The area of Antarctica used to calculate the continent-wide flux was  $14 \times 10^6 \text{ km}^2$ <sup>32</sup>.

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## Acknowledgments

The data reported in this paper are tabulated in the Supplementary Information. This work primarily was supported by the U.S. National Science Foundation Division of Polar Programs (research grants 9903744, 0538427, 0538416, 0968391, 1142166, 0632031; instrument grants 0216552, 0421412). We gratefully acknowledge the national research programs for their support and the efforts of field teams and others for help in collecting the ice cores in the array. The authors also thank the WAIS Divide Science Coordination Office for the collection and distribution of the WAIS Divide ice cores, Ice Drilling and Design and Operations for drilling support, the National Ice Core Laboratory for curating many of the cores used in this study; Raytheon Polar Service for logistics support in Antarctica; and the 109<sup>th</sup> New York Air National Guard for airlift in Antarctica.

## Author contributions

J.R.M., O.J.M., M.S., L.L. and R.E. conducted the ice core measurements at the Desert Research Institute. J.R.M., H.A., S.K., E.T., S.B.D., R.C.B., T.N. and M.A.J.C. provided ice core samples and assisted in the interpretation. J.R.M., O.J.M. and P.V. wrote the manuscript. All authors reviewed the manuscript.

## Additional information

**Supplementary information** accompanies this paper at <http://www.nature.com/scientificreports>

**Competing financial interests:** The authors declare no competing financial interests.

**How to cite this article:** McConnell, J.R. *et al.* Antarctic-wide array of high-resolution ice core records reveals pervasive lead pollution began in 1889 and persists today. *Sci. Rep.* **4**, 5848; DOI:10.1038/srep05848 (2014).



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